Asian aerosols in North America: Extracting the chemical composition and mass concentration of the Asian continental aerosol plume from long-term aerosol records in the western United States

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[1] Empirical assessment of the frequency and intensity of dust transport from Asia to North America has found that the dust regularly impacts elevated sites in the western United States, revealing a pattern of consistent, frequent transport above the marine boundary layer. Using the dust as a marker for Asian transport, a subset of Asian-influenced samples was identified within a decade of routine aerosol samples from two sites in the western cordillera of North America: Crater Lake, Oregon, and Mount Lassen, California. This subset was used to guide a statistical analysis to isolate Asian aerosol against the "background" of local contaminants. The analysis was then generalized to all samples during the transport season (March–October) for 1989–1999. A mixture of dust and combustion products dominates the Asian aerosol with typical concentration around 5 $\mu g/m^3$ and mass median diameter between 2 and 3 μm. Major fine particle (<2.5 μm diameter) constituent fractions are ~30% mineral, 28% organic compounds, 4% elemental carbon, 10% sulfate, <5% nitrate, and <1% sea salt. A second, possibly Asian, component of aged biomass smoke and sea salt is also present, with typical concentration (when present) around 1.5 $\mu g/m^3$. Averaged over the transport season the dusty Asian aerosol and the smoky aerosol comprise 60 and 6%, respectively, of total particle mass (<10 μm diameter) and 72 and 13% of fine particle mass at these sites. These data indicate that the Asian continental plume is a significant contributor to aerosol loading at remote high-altitude sites across western North America. This implies a significant influence for Asian emissions on free troposphere aerosols over North America and suggests that they need to be explicitly accounted for in aerosol analyses ranging from climate studies to aerosol regulatory programs. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0322 Atmospheric Composition and Structure: Constituent sources and sinks; 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; *KEYWORDS:* troposphere, aerosol, North America, Asia

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1. Introduction

[2] Atmospheric transport of natural and anthropogenic contaminants into and across the North Pacific basin can impair health and welfare in populated regions of the western Pacific, impact biologic systems throughout the northern Pacific, impact climate on ocean basin to hemispheric scales, and has the potential to impact air quality in North America [*Wilkening et al.*, 2000]. Gaseous and aerosol combustion products have been observed to accompany Asian dust at several sites within the Pacific basin, including Hawaii, Eniwetok, Midway, and others [*Shaw*, 1980; *Duce et al.*, 1980; *Jaffe et al.*, 1997; *Perry et al.*, 1999]. Limited monitoring on the Pacific coast of North America has detected transpacific transport of gaseous Asian tracers and pollutants, including ozone, carbon monoxide, nitrogen species, and organic compounds, as well as aerosols [*LeRoulley and Danielsen*, 1990; *Parrish et al.*, 1992; *Jaffe et al.*, 1999; *Tratt et al.*, 2001; *Kinoshita et al.*, 1999; *Uno*, 1999; *Berntsen et al.*, 1999; *Husar et al.*, 2001; *Jaffe et al.*, 2001].

[3] More recently, estimates of the long-term frequency and mass concentration of Asian dust transport have been reported for sites across North America, based on analysis of a decade of routine aerosol data [*VanCuren and Cahill*,

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2002]. Similar data on the nondust components of the Asian continental aerosol plume are needed to fully characterize that plume and to provide a basis for evaluating the Asian contribution to Northern Hemisphere tropospheric aerosols. This paper presents a statistical analysis of the nondust material that accompanied the Asian dust as observed in hundreds of aerosol samples collected between 1988 and 1999 and concludes that the Asian continental aerosol plume is a major contributor to tropospheric aerosols as measured at high-altitude ground stations in western North America.

[4] The findings reported here indicate that Asian aerosols are a regular component of the North American tropospheric aerosol with average concentration of roughly 5 $\mu g/m^3$ particulate matter less than 10 mm diameter (PM10), including $\sim$3 $\mu g/m^3$ particulate matter less than 2.5 mm diameter (PM2.5). This persistent aerosol impact is important geophysically because is suggests possible Asian influence on North America's continental radiation balance. It is also of interest for air quality regulation, as it constitutes about one fourth of the health-based annual PM10 and PM2.5 particle standards recently adopted in California [*California Air Resources Board* (*CARB*), 2001] or about one tenth and one fifth, respectively, of the health-based annual PM10 and PM2.5 standards recently adopted for the United States [*U.S. Environmental Protection Agency* (*USEPA*), 1997] and thus merits explicit consideration in regional aerosol analyses.

2. Analytical Approach

[5] Individual transport events as reported in the studies referenced above show transpacific transport to be highly variable. This study complements the detailed investigations by looking beyond individual events to report statistics for hundreds of transport events; the goal is to report aerosol climatology, not to explain variation among individual events. Working at a scale of continents and ocean basins, the mineral dust and combustion aerosols are assumed to have a common general geographic source area (East Asia). This analysis begins by using dust transport as a marker for Asian transport. Building on understanding of dust-marked transport, additional analyses are presented without that constraint. The results are a statistical representation of a decade of data showing the long term chemical composition, transport frequency, and mass concentration of anthropogenic pollutants transported across the North Pacific Ocean.

[6] A previous study used a distinct chemical signature derived from known Asian dust events to detect Asian dust in the long-term records of the Interagency Monitoring of Protected Visual Environments (IMPROVE) aerosol network [*VanCuren and Cahill*, 2002]. (For details of the IMPROVE program, see *Cahill and Wakabayashi* [1993] and *Pitchford and Scruggs* [2000].)

[7] The soil analysis approach cannot be repeated for combustion-derived pollutants since Asian combustion products neither can be shown a priori to be distinct from North American ones, nor are there any known overwhelming Asian transport events identified in the IMPROVE record to use to develop an in situ chemical profile for Asian combustion aerosols. Without an initial key to recognizing Asian combustion aerosols in the IMPROVE samples, the problem of identifying and quantifying combustion materials thus also entails resolving potential contamination with North American combustion aerosols.

[8] Despite these complications, a generally similar strategy can be used. Identifying the Asian soil material "marker" in IMPROVE samples required finding samples (events) in which the dust was demonstrably "pure" Asian dust. Characterizing the whole aerosol begins with looking at the nondust portion of samples containing the Asian dust marker then following one of two approaches: (1) Identify whole aerosol samples that are demonstrably "pure" Asian material or (2) infer the Asian composition in contaminated samples by statistically separating the Asian material from North American pollutants.

[9] The analysis begins by characterizing a subset of samples from selected sites (Crater Lake, Oregon, and Mount Lassen, California) then generalizing the analysis to assess the complete long-term record at these sites. The presentation follows this stepwise approach: section 3 covers the logic of the analysis and the selection of the study sites, section 4 demonstrates that these sites have substantial nondust Asian aerosol signatures and little potential contamination from continental North American emissions, section 5 examines the problem of recognizing and compensating for intraregional contamination by local emissions, section 6 presents a statistical view of the composition and prevalence of the Asian continental aerosol plume at these sites, and, finally, section 7 summarizes the meaning of these findings regarding the sources and transport dynamics of the Asian continental aerosol plume.

3. Obtaining an Asian Sample

3.1. Selecting Sites With Maximum Asian Exposure and Minimum North American Exposure

[10] Identifying sampling sites where events can be argued a priori to contain pure (approach 1) or minimally adulterated (approach 2) Asian material begins with the logic used in other studies of transoceanic transport: pick sites that appear to be upwind of all local sources. This is the basis for siting the University of Washington site at Cheeka Peak, Washington, on the Olympic Peninsula [*Jaffe et al.*, 1999] and the past and present United States National Oceanic and Atmospheric Administration Climate Monitoring and Diagnostics Laboratory (NOAA/CMDL) research sites at Point Arena and Trinidad Head, California [*Parrish et al.*, 1992, 2001]. This is also the logic of the siting of other globally significant "background sites" such as Mace Head, Ireland (http://www.igac.noaa.gov/newsletter/18/macehead.php). Coastal sites are desirable for intensive field campaigns supported by detailed meteorological analysis, as they permit sampling unmodified oceanic air masses selected for their transport history. By this logic, Point Reyes (Figure 1) would be the preferred IMPROVE site for total aerosol analysis.

[11] Unfortunately, the IMPROVE records analyzed here lack such support, so there is no basis to easily select particular samples that represent transport events known to have originated in Asia. Without detailed meteorological

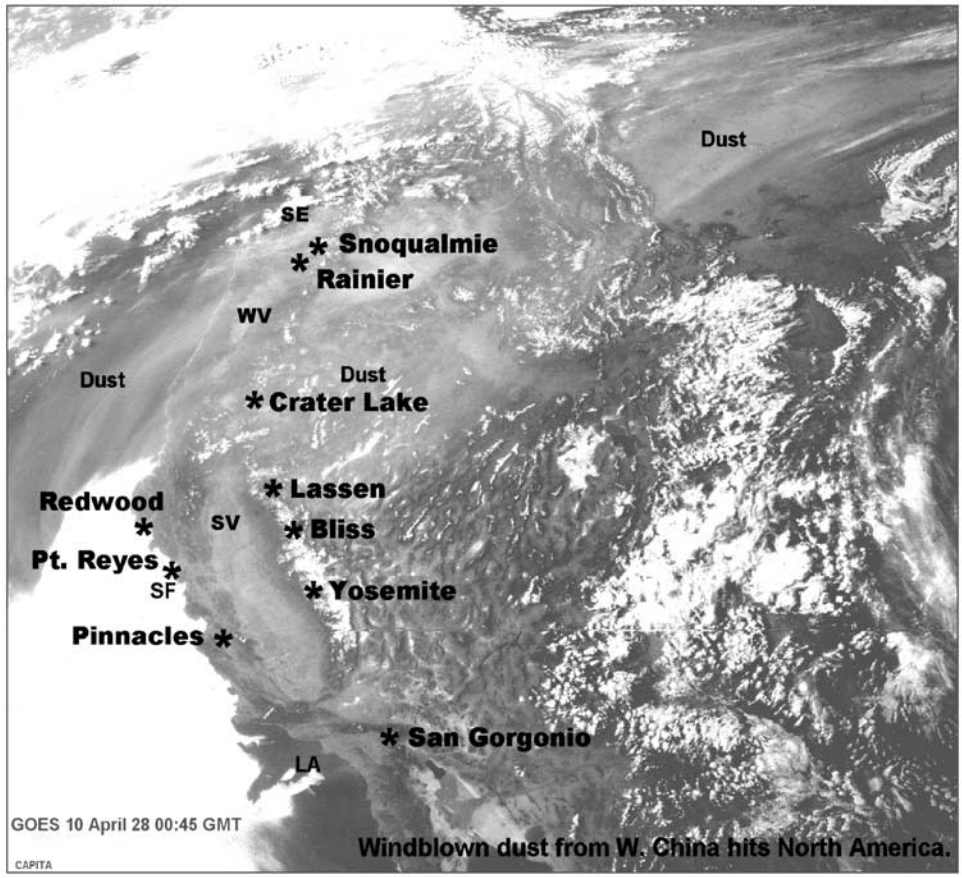

Figure 1. Geostationary Operational Environment Satellite 10 (GOES 10) visible band image of an Asian dust cloud passing over western North America. Stars are coastal and western cordillera IMPROVE sites discussed in text. Marine inversion is marked by low stratus clouds shrouding central and northern California coast; snow cover highlights Sierra Nevada crest and volcanic peaks of the Cascade Range. SE, Seattle; SF, San Francisco; LA, Los Angeles; SV, Sacramento Valley; WV, Willamette Valley (image modified from *Husar et al.* [2001]). See color version of this figure at back of this issue.

data, sea level sites on the Pacific coast of North America are ill-suited to sampling for transoceanic transport due to the strong and persistent marine inversion overlying the cold California current [*VanCuren and Cahill*, 2002]. Compromising between the ''absolutely coastal'' siting approach and the desire to have data from a site with observed robustness of Asian exposure leads to selecting a montane site in the western cordillera. These sites are generally above the marine boundary layer, but analysis of their records is complicated because, being somewhat inland, they may also be exposed to North American continental pollutants.

3.2. Recognizing Continental-Scale North American Influence

[12] To observe the strongest possible signal for the Asian continental aerosol plume, the selection criteria among sampling sites are high Asian dust frequency and minimal North American pollutant exposure. This leads to a simple logical model for this task: (1) Select sites with Pacific exposure and high Asian dust frequency (Figure 2, left). (2) Since decadal mean concentrations for Asian dust are relatively evenly distributed (Figure 2, right), we temporarily assume that other Asian contaminants over North America are also evenly distributed. (3) It follows that a monitoring site's mean concentration of a pollutant common to Asian and North American sources is the sum of a spatially invariant (or weakly variable) Asian component and a spatially variable North American component. Mathematically,

$$\overline{C}_t = \overline{c}_1 + \overline{c}_a,$$

where C_t is total observed concentration, and c_1 and c_a are the local and Asian partial concentrations, respectively. (In this context, ''spatially invariant'' refers to mean concentrations at well-exposed sites. Individual events often show very large spatial variation, so this model is applicable only to means.) (4) To choose among sites with similar frequency of Asian exposure, statement 3 implies that samples from the site with the lowest long-term mean contaminant concentrations contain the least North American material. (5) Since it is possible that there are unknown processes influencing individual pollutant species concentrations at the selected site(s),

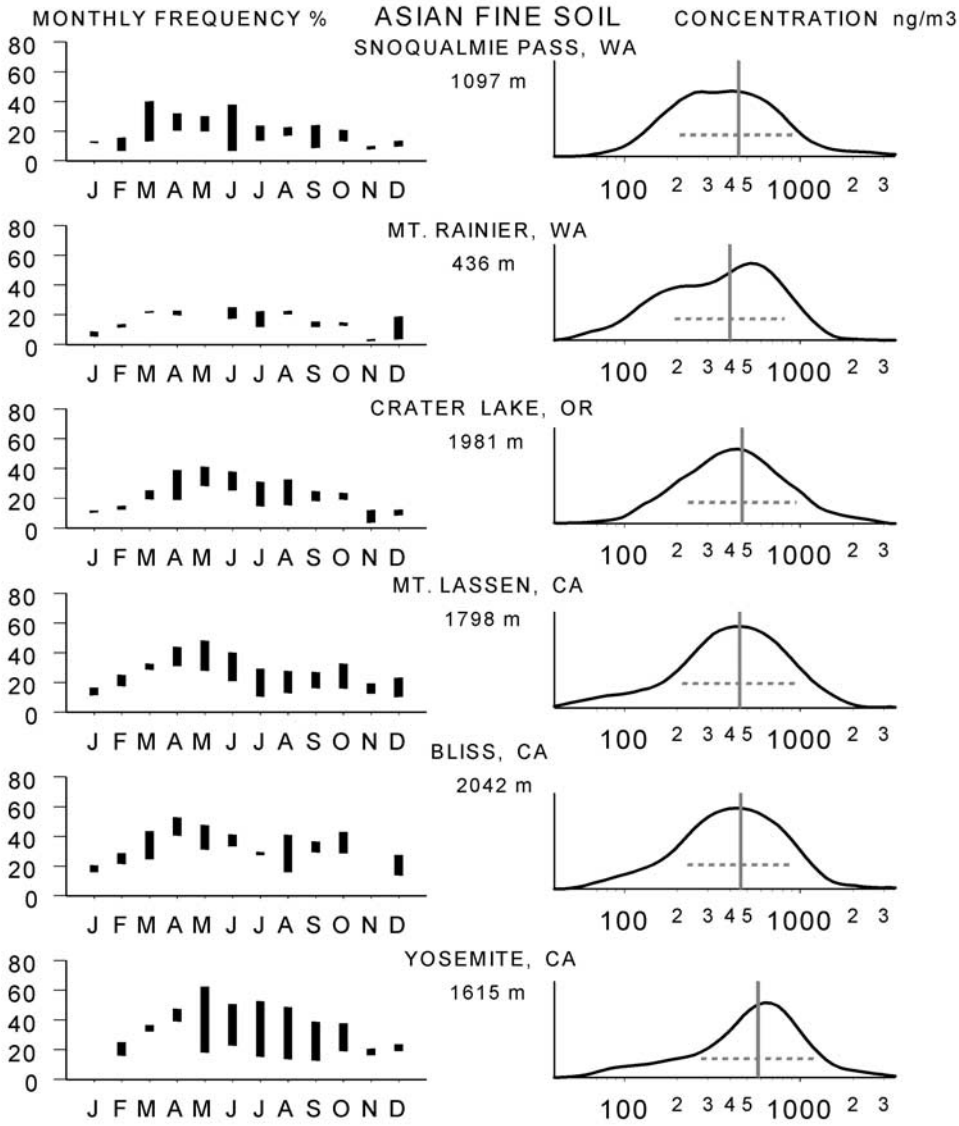

Figure 2. Asian dust monthly frequencies (range of low and high estimates from *VanCuren and Cahill* [2002]) and 10-year fine soil mass distributions (ng/m^3) for elevated sites in the western cordillera of North America. Lines (right) denote geometric means (vertical bars) and standard deviations (horizontal extent). Strong concentration consistency across sites confirms the assumption of low spatial variability for Asian impacts at elevated sites.

screening for more than one pollutant is used since agreement across multiple pollutants increases confidence in the results.

4. Using Urban/Industrial Marker Species to Rank North American Impact at IMPROVE Sites

[13] An ideal species for application of the minimal impact selection test would be a chemical that is inert (so that it does not get removed by reaction or deposition), unique to urban/industrial emissions (entirely anthropogenic), produced in both Asia and North America, easily detected, and widely measured and reported. Since fuel use is a dominant source of air pollutants in urban and industrial settings, likely candidates should be associated with energy production and consumption. *Holdren and Smith* [2000] provide global estimates of energy-related air pollutant production that suggest two possible indicators: lead for urban and transportation emissions and sulfur for urban, industrial, and electric utility emissions. Neither meets the ideal specification, but both have significant utility for this analysis. In the following sections each is considered and, with appropriate qualifiers, applied to the problem of site selection for Asian aerosol analysis.

4.1. Lead

[14] Lead meets the definition of a predominantly anthropogenic pollutant. World Energy Council data show that leaded gasoline and lead processing together account for ~95% of lead in the atmosphere [*Holdren and Smith*, 2000]. Globally, ~40% of airborne lead is directly emitted from motor vehicles by combustion of leaded gasoline and

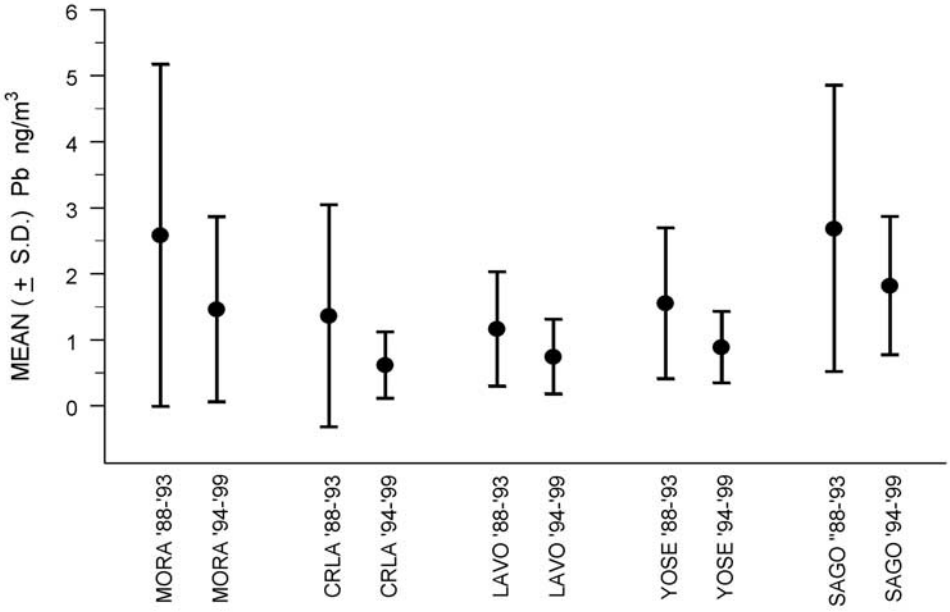

Figure 3. Changes in mean Pb concentrations at Cordilleran IMPROVE sites as a result of the phaseout of leaded gasoline. Crater Lake has the lowest current Pb concentrations, but smaller dispersion in ''prephaseout'' data at Mount Lassen suggests a smaller effect at that site. This difference does not appear to be an effect of local traffic visiting these national parks (Figure 4). MORA, Mount Rainier; CRLA, Crater Lake; LAVO, Mount Lassen; YOSE, Yosemite; SAGO, San Gorgonio.

as an erosional product from exhaust systems of vehicles previously operated on leaded gas; the remaining 60% comes from lead smelting and fabrication (especially batteries), waste incineration, and road dust contaminated with exhaust lead [*Holdren and Smith*, 2000]. In North America today, only ~16% of lead emissions comes from motor vehicles, while ~75% comes from industrial and waste disposal activities [*USEPA*, 2000]. Lead oxides and inorganic salts are nonreactive primary pollutants so that lead concentrations in source areas (e.g., cities) are quantitatively related to emissions. Far downwind from urban areas, for example at IMPROVE sites, lead concentrations have historically been used as indices of urban and transportation source impacts.

[15] Lead analysis in this case, however, is complicated by the demise of leaded gasoline in North America. Lowering North American emissions will have caused the relative intensity of the Asian continental lead signature to increase over time relative to the North American one, thus, although lead is a strong candidate as a marker for Asian material, long-term lead statistics may violate the assumptions supporting the minimum concentration criterion.

[16] Fortunately, the lead reduction itself can be used to directly compare IMPROVE sites' relative exposure to North American lead emissions. North American lead emissions peaked at ~225,000 t/year in 1972 then declined to <5000 t/year by 1998 [*USEPA*, 2000]. The bulk of this reduction came from the removal of lead from gasoline, a process completed by 1992. Figure 3 shows the response in measured lead concentrations at the cordilleran IMPROVE sites. It is clear that the emission reductions reduced lead concentrations at all sites; moreover, the trend toward larger changes at sites with higher mean concentrations (Mount Rainier and San Gorgonio) indicates that emission changes in large metropolitan areas (Seattle and Los Angeles, respectively) are reflected in these data.

[17] The lead data suggest that Crater Lake and Mount Lassen have the least North American urban exposure, but, since lead is strongly linked to motor vehicles, local traffic effects need to be considered as well. Figure 4 shows the annual trends of visitation and lead concentrations before and after the completion of the lead phaseout. It is clear from these data that local traffic is not controlling lead concentrations at either Crater Lake or Mount Lassen, and the lowest mean concentration selection criterion is applicable to these sites.

4.2. Sulfate

[18] Globally, nearly one fourth of airborne sulfur is of oceanic origin, while three fourths of atmospheric sulfur comes from human activity; ~85% of anthropogenic sulfur comes from burning fossil fuels [*Holdren and Smith*, 2000]. Aerosol sulfur is robustly measured by the IMPROVE protocol, both as an element (proton-induced X-ray emission (PIXE) analysis) and in the form of sulfate (ion chromatography). The fact that montane IMPROVE sites are generally exposed to the free troposphere and largely isolated from oceanic aerosols suggests that sulfate may fit the model described above as a marker for fossil fuel combustion in data from the montane cordilleran sites. North American sulfur emissions were reduced over the decade of the 1990s, but unlike lead, total emissions remained strong in North America over the period of the IMPROVE data [*USEPA*, 2000]; on the basis of emissions alone, the minimum concentration criterion is valid for sulfur.

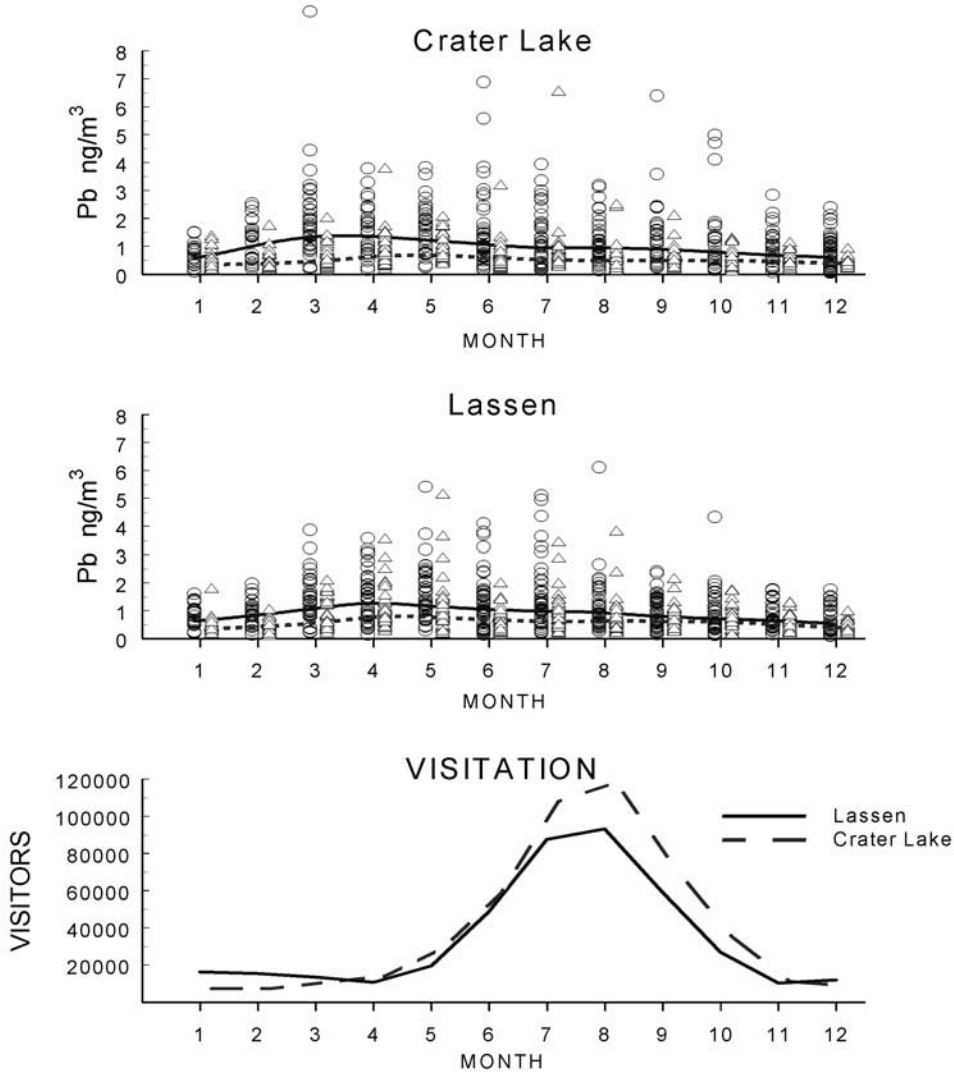

Figure 4. Seasonal distribution of Pb concentrations and visitation at Crater Lake and Mount Lassen before and after the phaseout of leaded gasoline in North America. Symbols are Pb concentrations sorted by month, and curves connect monthly means. Circles and solid line denote late phaseout (1990–1992); triangles and dashed line denote after phaseout (1995–1996). Both sites exhibit a springtime peak, out of phase with the July–August peak in visitation (bottom). Lead concentrations are not strongly related to local activity at either site (Visitation data from National Park Service Web site, available at http://den2-11.den.nps.gov/stats/).

[19] There are, however, other substantial problems with using sulfate as a marker. Sulfur emissions are not a linear function of source area fuel use within North America due to regional differences in sulfur content of fuels. In addition, aerosol sulfate is not a quantitatively stable product of combustion due to the highly nonlinear process of conversion of gaseous sulfur emissions into sulfate. Finally, sulfate is common in desert dust (a likely component of Asian aerosols) and may be incorporated into the Asian plume from the ocean during transport; while these factors do not disqualify sulfate as a marker for transport, they suggest caution in its use as a tracer of anthropogenic material from Asia. Given these confounding factors, the potential for sulfate to violate the assumptions of the minimum mean concentration criterion must be considered.

[20] The nonlinearity of sulfate formation makes individual transport events highly idiosyncratic; however, long-term statistics are not necessarily so severely compromised, since the conditions of transport, temperature, humidity, residence time, etc., are not independently variable. Just as mean climate statistics are more stable than daily weather, transport processes tend to be replicated over time as well. This point is illustrated by the plots in Figure 5. The urban data (Lynwood versus Rubidoux) are presented to show how sulfate statistics behave in a case of clear source-receptor association (Los Angeles megalopolis). Although the controlling transport factors are complex, repetitious meteorological conditions (i.e., ''climate'') tend to make concentrations in the upwind source area (Lynwood) and at the downwind site (Rubidoux) strongly, if noisily, corre-

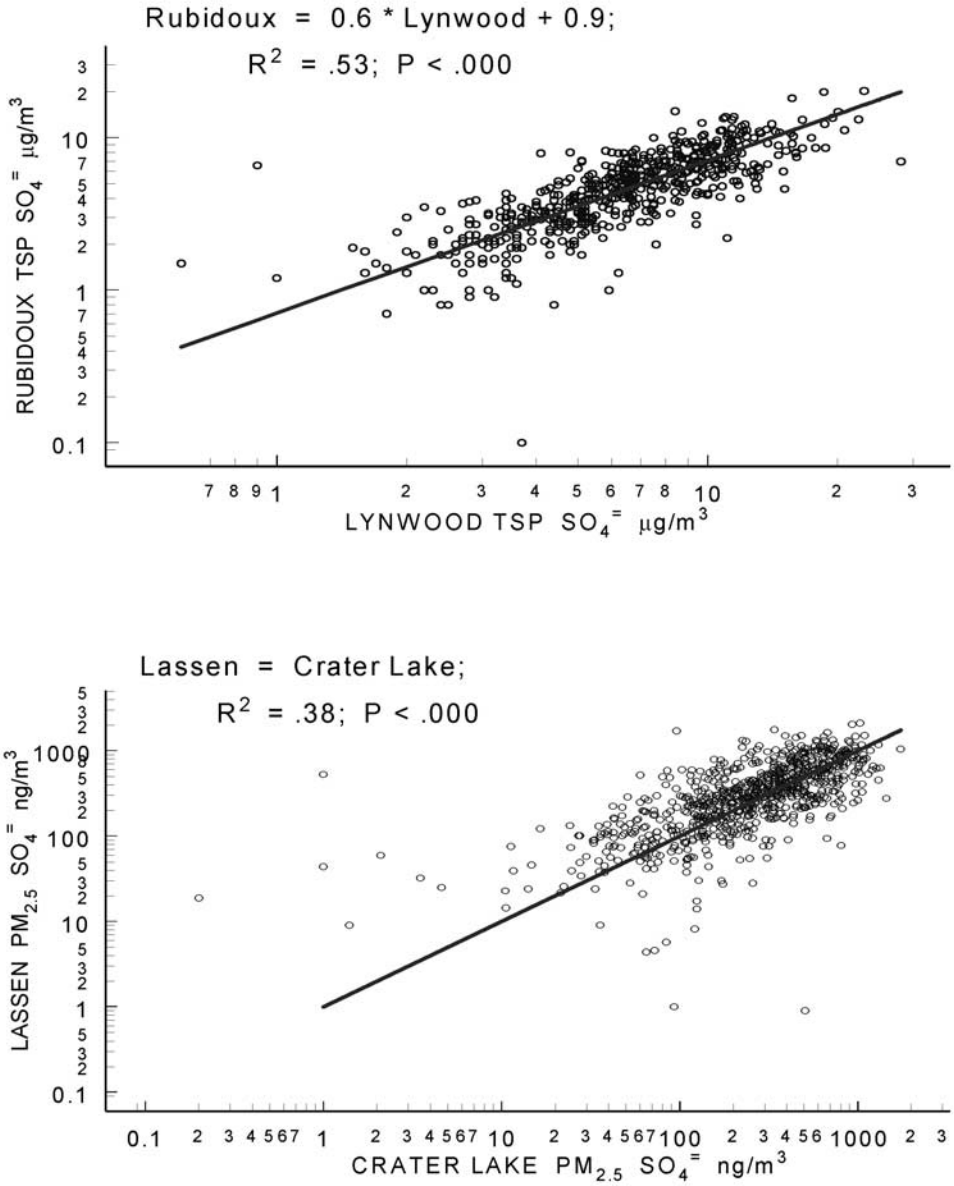

Figure 5. Spatial repeatability of sulfate formation and transport. Regression of 24-hour sulfate concentrations on paired days. Urban case (Los Angeles megalopolis), Rubidoux, 77 km east of Lynwood, is nonetheless strongly linked to processes in the upwind metropolitan areas as represented by Lynwood data [CARB, http://www.arb.ca.gov/regact/aaqspm/res0224.pdf, 2001]; this case represents typical processes responsible for the North American sulfate contribution at IMPROVE sites. Rural case, Mount Lassen and Crater Lake, although separated by 265 km, experience the same regional air masses much of the time and thus share a common sulfate exposure history; this case shows the spatial stability of sulfate transport processes. Together, these data show that complex sulfate chemical kinetics tend to repeat in time and space at local to hemispheric scales; thus they do not preclude simple statistical association.

lated. On a continental to hemispheric scale, the same stability of processes is reflected in the regression of paired day data at Crater Lake and Mount Lassen. Despite being separated by hundreds of kilometers and exposed to different local sources, both sites experience essentially the same regional sulfate ''climate.'' Given this repeatability at local to continental scales, sulfate kinetics can be treated as a ''black box'' in this analysis, and the minimum mean concentration criterion can be applied to sulfate data.

[21] Sulfate frequency distributions for all sampling days for the cordilleran IMPROVE sites are shown in Figure 6. All the sites share a peak near 100 ng/m^3, but there is considerable variation at higher values. This is the pattern predicted by the minimum concentration concept developed above. The plots clearly show that Crater Lake and Mount Lassen experience the least North American sulfate exposure. In fact, the lack of strong secondary peaks suggests that these sites come close to satisfying the initial goal of

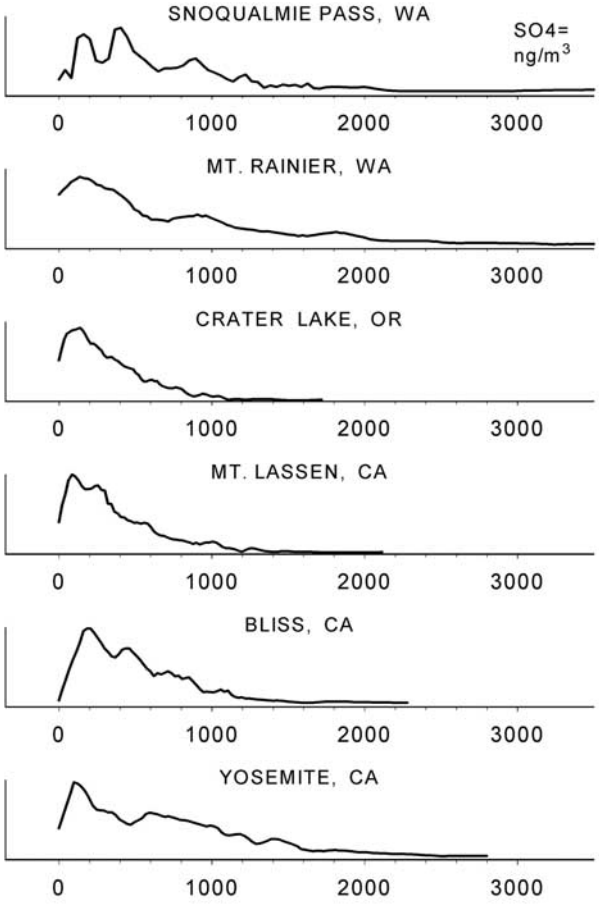

Figure 6. Frequency distributions for aerosol $SO_4^=$(ng/m^3) for IMPROVE sites along the western cordillera of North America ($n \cong 1000$ per site). Most sites exhibit a peak at the regional minimum, roughly 100 ng/m^3. This appears to represent the spatially invariant mean Asian fraction. Site-specific secondary peaks reflect the diverse patterns of intracontinental North American sulfate transport. Crater Lake, and to a lesser degree Mount Lassen, display low modal values and compact distributions consistent with the criterion of minimum local North American influence.

finding sampling sites that are upwind of all local sources (approach number 1, section 2).

5. Aerosol Composition at Crater Lake and Mount Lassen

5.1. Evidence of a Common Strong Source

[22] The Asian soil data (Figure 2) show that there is a broadly uniform Asian aerosol impact across the western United States. The lead and sulfate data indicate that North American sources have relatively small impacts at Crater Lake and Mount Lassen, and the sulfate data also suggest that the Asian signature at these sites is strong relative to any North American one. This leads to a first-order interpretation of the whole aerosol accompanying Asian dust at these sites as being primarily Asian. Figure 7 compares the mean percent mass fractions of major chemical species on days identified as containing Asian dust at these sites. The strong similarity of the compositions suggests a common source, and the composition, high in sulfate, nitrate, and carbonaceous aerosol, as well as dust, indicates a significant anthropogenic component. This level of analysis indicates that the gross composition data for these sites when Asian dust is present approximates the general character of the Asian continental aerosol plume, but it leaves open the following three questions: (1) How much local material (what could be characterized as the montane ''background'') is present on days when Asian dust is present? (2) Are there days when Asian aerosols are present but the dust fraction is not detectable by these methods? (3) Are there days when Asian aerosols are present but the dust is absent?

5.2. Local Contamination: Biomass Smoke

[23] Although there do not appear to be significant urban and industrial aerosol sources affecting Crater Lake and Mount Lassen, there are still significant local aerosol emissions. Crater Lake and Mount Lassen are surrounded by vast stretches of forest, creating several sources of biomass smoke. Wildfires are common in the region. Infrequent large fires spread smoke over thousands of square kilometers, while numerous small fires constitute a more persistent regional-scale seasonal aerosol source. Commercial timber production creates emissions from harvest waste (''slash'' burning), controlled burning to manage forest structure (''prescribed fire''), and the use of mill waste as boiler fuel. Wood is a common domestic heating fuel in rural communities in the region and recreational campfires are common during the warm months. In addition, grain and grass seed farming in surrounding lowlands occasionally contributes agricultural smoke. Because these intraregional sources are in the vicinity of the sampling sites, the general isolation from North American urban and industrial aerosol sources identified above does not apply to their emissions.

[24] Evaluating the biomass contribution to aerosol loading at these sites requires a chemical marker for smoke that distinguishes it from the Asian aerosol. This was solved using a ''found'' experiment (i.e., an event in the record that permits an unambiguous characterization of local wildfire smoke). In July 1999 a lightning storm across extreme Northern California and Southern Oregon ignited hundreds of fires [*Bryant*, 2000]. Most of the fires were quickly extinguished, but one, the Megram/Big Bar fire in the Trinity Alps Wilderness Area, ~160 km (100 miles) WNW of Mount Lassen, continued into November, eventually burning more than 50,000 hectares (125,000 acres). During that fire's lifetime its smoke plume variously flowed south and east, providing a time series of alternating clear days and extreme smoke events at Mount Lassen.

[25] Comparing clear and smoky samples from Mount Lassen and comparing samples from Crater Lake and Mount Lassen on paired days shows that the carbon/sulfate ratios in the samples are strongly dependent on the fraction of smoke in the samples (Figure 8). Ratios for nonsmoky days exhibit a mean of 2.6 ± 2.0, while strong smoke events' ratios ranged from 12 to 60 with a mean near 30.

[26] The Big Bar fire C/$SO_4^=$ ratios are presented along with several published smoke profiles for the region in Table 1. Frequency distributions of carbon/sulfate ratios on

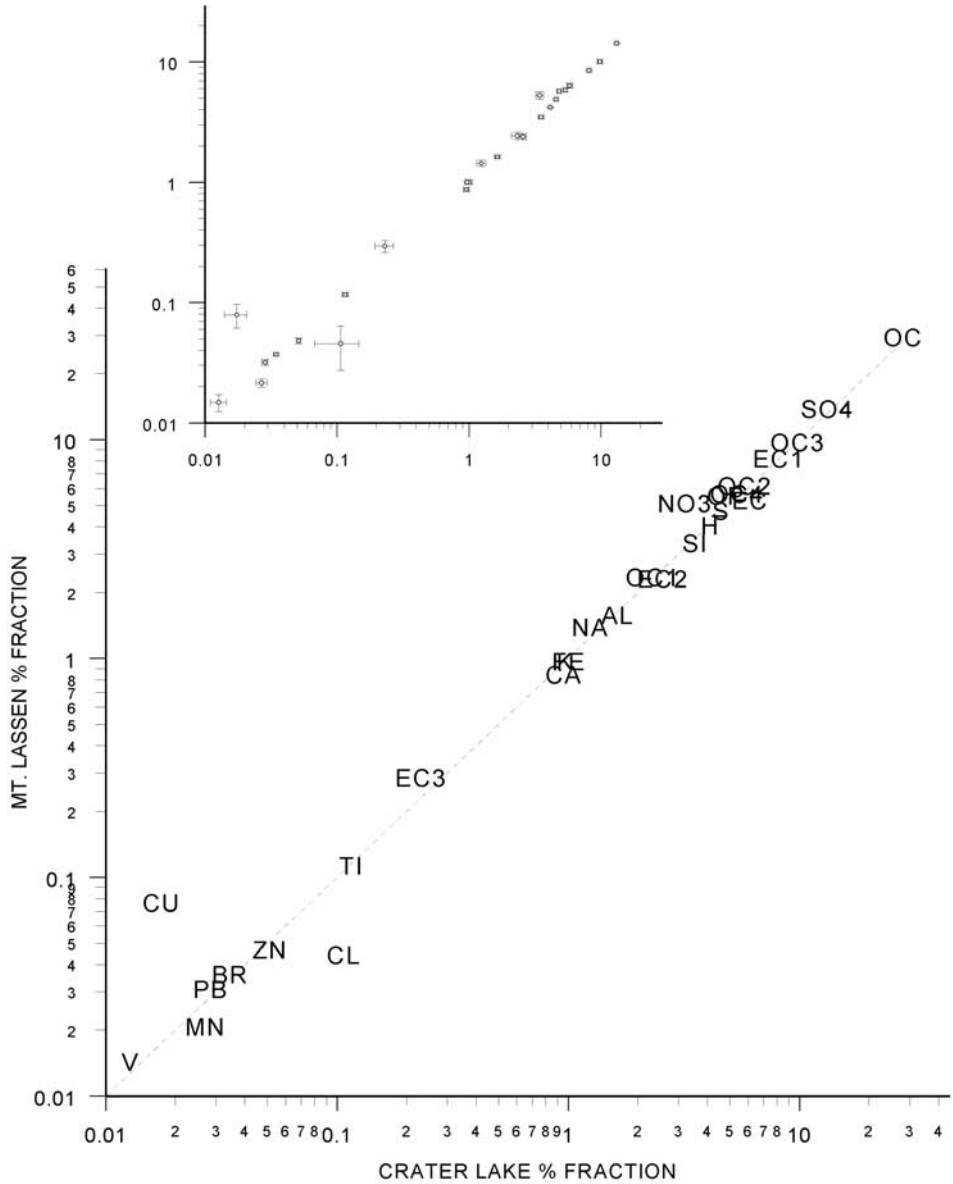

Figure 7. Scattergram comparing mean fraction of fine mass by species for days with Asian dust at Crater Lake and Mount Lassen showing very close agreement on the whole aerosol composition. Means computed for 258 days at Crater Lake, 349 days at Mount Lassen. Overlapping species are K and Fe, OC1, and EC2. Species shown account for more than 99.9% of analyzed sample mass. Copper is a known artifact at Mount Lassen, and chlorine is below detection limits in more than three fourths of samples (nondetection values set to zero in calculating mean). Dashed line is 1:1. Inset shows standard errors of means.

days with high-confidence Asian dust identifications for the entire data set (259 samples at Crater Lake, 349 at Mount Lassen), and a simple mixing model based on these data is shown in Figure 9. The shapes of the frequency distributions suggest two independent but overlapping distributions: one for the Asian aerosol, the other for biomass smoke. The low-smoke mode ($C/SO_4^=$ ratios below 6) contains ~90% of the samples, while moderately smoky days ($C/SO_4^=$ ratios in the range 6 to 15) extend to the 97th percentile, and severely smoky days (ratios above 20) are restricted to the top 1% of samples. The mixing model indicates that the smoke fraction of total carbon is less than half for $C/SO_4^=$ values below 5, resulting in a fine mass smoke fraction below one fourth; at ratios below 3 the smoke carbon fraction is <0.1 and smoke mass fractions are very small.

5.3. The Low-Smoke/High-Confidence Asian Dust Sample Set: A First Estimate of the Composition of the Asian Continental Aerosol Plume

[27] In an effort to compromise between capturing the range of composition in the Asian aerosol and minimizing smoke-induced bias, an initial Asian aerosol chemical characterization was calculated using pooled statistics for all samples with $C/SO_4^=$ ratios below 4. Table 2 shows the

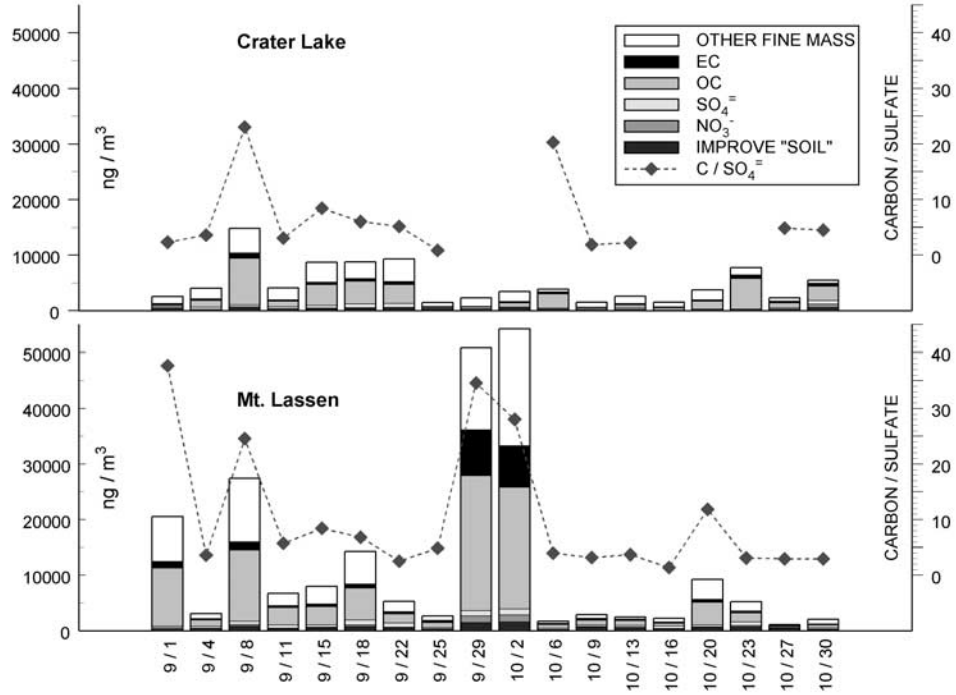

Figure 8. Time series fine aerosol major chemical species at Crater Lake and Mount Lassen, 1 September–30 October 1999. Eastern Pacific air masses with typical fine aerosol burdens of 2–6 μg/m^3 are interrupted by occasional wildfire smoke impacts. The carbon mass change dominates the smoke events, so smoke events exhibit characteristically high carbon/sulfate ratios. Low-smoke days show a lower bound carbon/sulfate ratio in the range of 3 to 6; smoky days regularly exceed 20 (Crater Lake carbon data missing for 29 September; sulfate missing for 2 October). See color version of this figure at back of this issue.

resulting mean composition of the Crater Lake - Mount Lassen regional aerosol for 392 sample days with low-smoke and high-confidence Asian dust.

5.4. Chemical Composition of the Asian Continental Aerosol Plume

[28] The data in Table 2 are a reasonable estimate of the composition of the Asian continental aerosol plume based on the logic of the minimum mean concentration site selection criterion and correcting for known local emissions. However, it may not correctly represent the Asian continental aerosol plume if (1) there are biases associated with nonsmoke periods, (2) there are other significant local sources active on these days, (3) the smoke mixing model is incorrect, (4) the assumption of minimal North American input is incorrect, or (5)the Asian plume has a dust-free phase.

[29] Nonetheless, the low-smoke/high-confidence Asian dust sample population is a solid starting point since it is clear that it is dominated by days with Asian influence. Thus it should at least be possible to retrieve an unequivocal signal for the dusty phase of the Asian continental aerosol plume.

[30] Progressively expanding the analysis to include smoky days and "nondust" days enables recognizing the effects of smoke, detecting strong bias connected with selecting subsets of days, and determining whether or not there is a dust-free phase to the Asian aerosol plume. Principal component analysis (MATLAB version 5.3, Mathworks, Inc., Natick, Massachusetts, 1999) was used to look within this data set to identify statistically significant aerosol types (in this case "aerosol type" means a suite of chemical species that vary together with temporal independence from other components of the bulk aerosol). The analysis used 13 IMPROVE species that are consistently above minimum detection in the Crater Lake and Mount Lassen data plus the following three derived values: K_{non} is the estimated nongeogenic (i.e., combustion) potassium; it is calculated by assuming that Al and mineral K are very highly correlated, then using the minimum observed K/Al

Table 1. Carbon/Sulfate Ratios for Several Published Smoke Profiles [*USEPA*, 2002], the Data From the Big Bar Fire, and the Entire Set of High-Confidence Asian Dust Days From Crater Lake and Mount Lassen[a]

Case	$C/S0_4^=$	Max	Min
Flaming conifer slash	18.6	33.2	12.9
Smoldering conifer slash	10.6	29.3	6.5
Medford slash	29.9	41.2	23.5
Big Bar fire	29.6	57.4	12.5
Prescribed conifer composite	429.9	671.0	316.3
Willamette rye grass	30.7		
Willamette fescue grass	19.9		
California field crop	10.8		
Medford residential	56.2	122.6	25.8
Pocatello residential	377.7	493.0	306.0
Smoke geometric mean	40	235	13
Mean and std. dev. Asian dust days at Crater Lake and Lassen	2.62	4.56	0.68

[a]Max, maximum; min, minimum; std. dev., standard deviation.

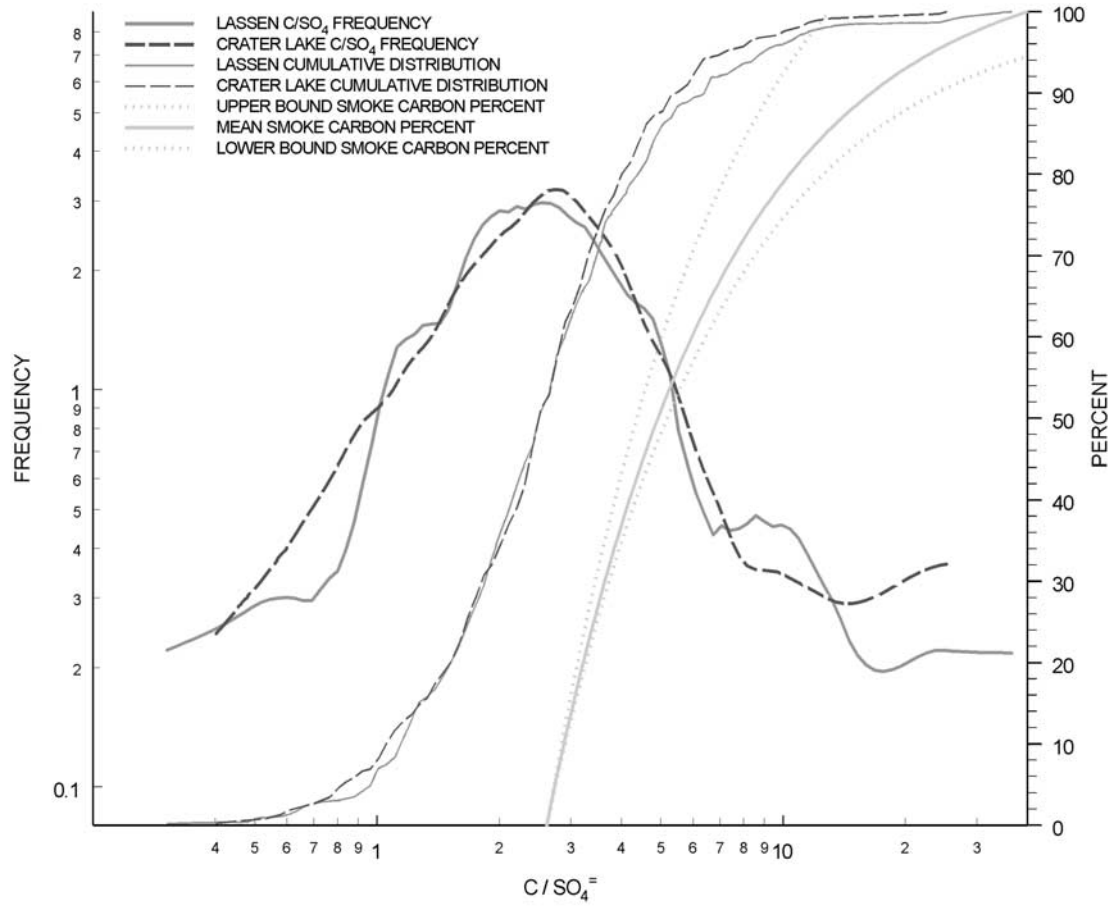

Figure 9. Frequency distributions of $C/SO_4^=$ ratios for days with high-confidence Asian dust scores at Crater Lake (258 days) and Mount Lassen (349 days) from 1989–1999. The low to moderate smoke mode ($C/SO_4^= < 6$) contains ~90% of the samples. Two smoke-influenced modes are also present, one in the range 6 to 15 (92nd to 97th percentiles), and a second above 20 (top 1% of events). The solid and dashed lines rising from the center show smoke carbon fractions calculated using a simple mixing model assuming the dusty Asian aerosol has a $C/SO_4^=$ ratio of 2.7 (the mode of the distribution) and the overall minimum, maximum, and mean smoke ratios from the line ''smoke geometric mean'' in Table 1. Smoke is generally less than half of the carbon and less than about a quarter of total fine mass in the sample population with ratios below 5. Analyses reported for ''low-smoke'' samples used pooled statistics for all samples below 4 as a compromise between capturing the range of Asian aerosol composition and excluding influence from smoke. See color version of this figure at back of this issue.

ratio in the data set to compute; K_{non} = K – (Al × 0.26). OC and EC are organic and elemental carbon from the IMPROVE thermal-optical reflectance (TOR) carbon data, calculated as OC = OC1 + OC2 + OC2 + OC4 + OP and EC = EC1 + EC2 + EC3 -OP [*Chow et al.*, 1993]. The K_{non} transformation permits using nonmineral potassium as a marker for biomass smoke, while the carbon simplification reduces carbon from eight variables to two, thus preventing carbon variances from dominating the analysis and allowing more direct comparison with published source profiles.

[31] The large differences in mass concentrations between species were balanced by converting the concentration values to z-scores (the quotient of the concentration difference from the species mean divided by the species standard deviation). The z-transform allows the analysis to find species assemblages that vary together, regardless of whether the species are major or trace constituents. The results are

Table 2. Gross Composition Statistics ($\mu g/m^3$) of Crater Lake and Mount Lassen Aerosol for 392 Sampling Days With High-Confidence Asian Dust and $C/SO_4^=$ Ratios Below 4[a]

Fraction	Mean	Std. Dev.
Total mass, PM10	7.9	5.3
Coarse mass, PM2.5–10	3.9	3.8
Fine mass, PM2.5	3.9	2.4
Total carbon	1.2	0.69
Organic carbon	0.98	0.59
Elemental carbon	0.18	0.11
Sulfate	0.56	0.33
Fine soil	0.65	0.95
Nitrate	0.17	0.21

[a]This approximates the dusty Asian aerosol as defined by the principal component analysis (PCA) presented in section 6. Chemical types are only available for the PM2.5 fraction in IMPROVE data. Note that the 2.5 μm cut approximates the mass median diameter in this data set.

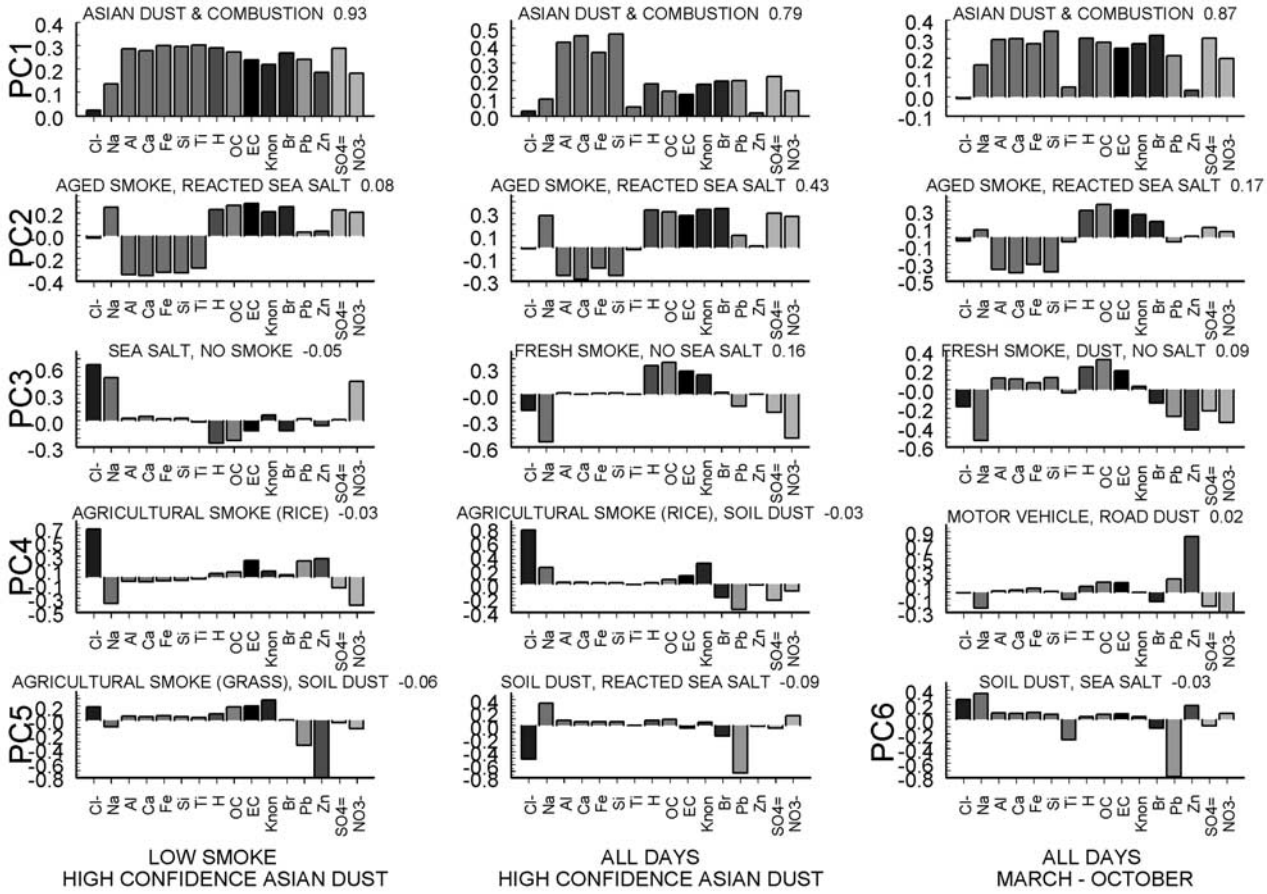

Figure 10. Leading Principal Components (PCA) of pooled IMPROVE data for Crater Lake and Mount Lassen 1989–1999. Left panels show 392 sample days with low smoke ($C/SO_4^{=} < 4$) and high-confidence Asian dust by method of *VanCuren and Cahill* [2002]. Middle panels show 501 sample days with high-confidence Asian dust. Right panels show 1234 sample days in the transport season (March–October). Components computed from measurements transformed to z scores (units of σ) to balance sensitivity among dominant and trace species. The PCs express the covariance structure of the correlation matrix to show which species vary together. Bars represent strength of covariance, not relative mass. Label on each plot gives subjective source category identification and the correlation coefficient of the component with fine aerosol mass. The dusty Asian plume (PC1) and aged smoke with sea salt (PC2) are dominant across all data sets. Agricultural burning and sea salt signify upslope transport. Moving from the selected dust data to dust with smoke to the whole transport season data set there is no new major category associated with the non-dust-identified days (more than half of all days), but weak local sources (fresh smoke, local dust, and vehicle exhaust) rise in importance. The transport season case includes many low-aerosol concentration days associated with the dusty Asian aerosol; thus mass correlation increases even with the strict dust constraint removed. (Transport season PC5, not shown, is an artifact due to nondetection of Zn on low aerosol mass days.) A regression-based mass model for these data is presented in Table 3. See color version of this figure at back of this issue.

presented graphically in Figure 10; notations in the plots give subjective source category assignments based on published source profile data [*USEPA*, 2002; *Jenkins et al.*, 1995] and correlation coefficients with fine aerosol mass.

6. Aerosol Composition on Days With Strong Asian Dust Signatures but Minimal Smoke

[32] The principal component analysis (PCA) results for 392 low-smoke/high-confidence Asian dust case (Figure 10, left) follow the pattern expected from the analysis in section 5.

[33] The first component combines strong and highly correlated soil elements (expected because these sample days were selected based on meeting strict criteria for containing Asian dust) with a broad suite of combustion products and secondary species. The combustion-related species accompany the dust because their variation in concentration is temporally correlated with the dust. Together, this constitutes the dusty phase of the Asian continental aerosol plume. This component has a correlation coefficient to total fine aerosol of 0.93. Clearly, this material dominates the aerosol for this subset of sampling days.

[34] The second component is a carbon-dominated aerosol containing chemical markers for biomass smoke, modified sea salt (combustion-derived nitric acid converts NaCl to $NaNO_3$), and sulfate (derived from both sea salt and combusted biomass sulfur). The presence of secondary chemical products (nitrate, sulfate) suggests that this material is several hours to several days old when sampled, while the sea salt indicates deep atmospheric mixing and/or sustained residence time over the ocean. The carbon and sulfur in this component are chemically consistent with the large fire regional smoke profiles discussed in section 5.2. Regional smoke can intermingle with the Asian dust because the transport season and the fire season largely overlap, but the PCA analysis distinguishes it from the Asian material because its concentration does not vary synchronously with the Asian material.

[35] The third component is partially reacted sea salt, inversely correlated with smoke. This component is anticorrelated with total fine aerosol concentration, suggesting that it is strongest on days when the Asian signature is weak or absent. This is consistent with the previously reported mutual exclusion of dusty Asian aerosol and coastal marine air [*VanCuren and Cahill*, 2002]. This component's average mass contribution is minimal.

[36] The fourth component, rich in chlorine and carbon, appears to be smoke from rice stubble burning in the Sacramento Valley (California). Rice stubble smoke (PM2.5) is roughly one fourth chlorine [*Jenkins et al.*, 1995], and back trajectories [HYSPLIT4, available at http://www.arl.noaa.gov/ready/hysplit4.html, 1997] for strong events (sample days with high PC3 scores) pass through the Sacramento Valley. It, too, is anticorrelated with total fine aerosol concentration. Again, the average mass contribution is minimal.

[37] The fifth component is somewhat ambiguous. On the basis of elevated chlorine (grass stubble smoke from the Willamette Valley (Oregon) is ~6–10% chlorine [*USEPA*, 2002]), association of strong events with unusually high coarse particle fractions, and the fact that the scores drop abruptly in late fall (onset of the rainy season), this is tentatively identified as agricultural dust and smoke, and/or slash burning.

6.1. Aerosol Composition on All Days With Strong Asian Dust Signatures

[38] The middle column in Figure 10 shows results of the 16-species PCA applied to 501 high-confidence Asian dust days at Crater Lake and Mount Lassen. Comparing the fine mass correlation coefficients for the entire dust-related data set with the low-smoke subset shows the source of the high $C/SO_4^=$ events to be the second component, with minor contributions by the third and fourth. The general pattern is the same as the low-smoke case; there is no apparent bias in representing the dusty phase of the Asian aerosol when deleting smoky days from the analysis. Liberalizing the smoke restriction adds a local fresh smoke component (PC3); agricultural smoke persists, and there is more upslope transport of soil dust (component 5), possibly due to the higher incidence of deep atmospheric mixing in this data set.

[39] Significantly, the large increase in fine mass correlation coefficient for the second component seems anomalous in light of the limited number of strongly smoky days added by this change from the low-smoke set (see Figure 9). The combination of generally low mass concentrations, the oceanic chemistry, and the necessary Asian transport association in this set of sample days suggests that at least some of this smoke may be transported from Asia. If the second component constitutes a separate, "smoky" phase of Asian emissions, it is not simply a dust-free case of the first component, since it lacks the dusty phase's strong motor vehicle markers of lead and zinc.

6.2. Aerosol Composition Throughout the Transport Season

[40] Figure 10 shows (left and center) unequivocally that combustion products accompanied the dust on the days previously identified as having Asian dust. However the use of dust to select the samples prevents the analysis from determining whether or not there may also be periods when the combustion products are present without the dust. Performing the same principal component analysis for all 1234 sample days in the transport season (March to October; see Figure 2) resolves this question (Figure 10, right). Several features stand out when comparing the results for the Asian dust subset with those for the whole transport season data set: Foremost, the general pattern is unchanged; the Asian dust and combustion aerosol is the dominant fine aerosol mode throughout the transport season. The association remains very strong even with the dust identification constraint removed, indicating that the dust and combustion products are intimately related. The fine mass correlation coefficient for the first component rises with the dust constraint removed, indicating that the dust is present even on days when the detection algorithm fails (low-mass days with poor quantification of some elements or days with local dust also present). Asian dust, at least in small amounts, appears to be present almost continuously through the transport season.

[41] The aged smoke component is slightly different in the transport season case, with a drop in the intensity of the reacted sea salt signature (Na, Br, $SO_4^=$, and NO_3^-). In addition, this component's fine mass correlation coefficient drops when the dust constraint is removed. This supports the inference that there may be a connection between aged smoke and Asian transport events.

[42] The third component, "fresh smoke, no sea salt" (center) gains an even stronger "local" character with the addition of local soil dust when the Asian dust identification criterion is removed (right). The temporal correlation of this dust and smoke indicates that they share a common emission pattern, strongly suggesting that this component is a result of daily National Park activity near the samplers (unpaved roads, campfires, etc.).

[43] Overall, in all three calculations presented in Figure 10, local sources such as road dust, campfires, and motor vehicle exhaust have minimal influence on fine aerosol mass. This rules out local activity as the source of the combustion aerosol at either site.

6.3. Characteristics of the Dusty Asian Aerosol

[44] The strong consistency of the signature for the dusty Asian aerosol across all three analyses allows use of a simple subset of days to provide a reasonable approximation

Table 3. PCA-Based Regression-Derived Concentrations ($\mu g/m^3$) of Major Aerosol Constituents by Source Category for Crater Lake and Mount Lassen[a]

	High-Confidence Asian Dust				Transport Season			
	Low-Smoke, C/SO4 < 4		No Large Local Fires, CSO4 < 6		All days March–Oct. 1989–1999			
	Mean	Std. Dev.	Mean	Std. Dev.	Mean	Std. Dev.	Mean, %	Std. Dev., %
Total mass, PM10	6.7	3.9	6.7	5.1	7.4	6.2		
Dusty Asian plume	6.4	4.2	4.9	4.3	4.5	4.0	60	54
Aged smoke and sea salt	0.15	0.10	1.7	1.2	0.46	1.2	6	16
Fresh smoke and local dust			0.02	0.25	1.5	1.7	20	23
Sea salt	0.02	0.26						
Road vehicle and dust					0.17	0.50	2	7
Agricultural smoke	0.06	0.88	0.01	0.45				
Sea salt and soil dust			0.04	0.67	0.93	0.93	13	13
Coarse mass, PM10-2.5	2.9	1.9	2.9	2.9	3.8	3.9	51	52
Dusty Asian plume	2.9	1.9	2.8	2.4	1.9	2.0	25	27
Aged smoke and sea salt	0	0	0	0.10	0	0.45	0	6
Fresh smoke and local dust			0	0.15	1.2	1.4	16	18
Sea salt	0.01	0.06						
Road vehicle and dust					0.17	0.47	2 6	
Agricultural smoke	0.05	0.81	0.01	0.05				
Sea salt and soil dust			0.04	0.57	0.88	0.86	12	12
Fine mass, PM2.5	3.8	2.0	3.8	2.3	3.6	2.4	49	32
Dusty Asian plume	3.5	2.3	2.1	2.0	2.6	2.0	35	27
Aged smoke and sea salt	0.15	0.10	1.7	1.1	0.46	0.74	6	10
Fresh smoke and local dust			0.02	0.10	0.26	0.39	4	5
Sea salt	0.02	0.20						
Road vehicle and dust					0	0.03	0	0
Agricultural smoke	0.01	0.07	0	0.41	0.07	0.18		
Sea salt and soil dust			0	0.11	0.05	0.07	1	1

[a]Results presented for 392 sampling days with low-smoke and high-confidence Asian dust (left), 459 sampling days with low to moderate smoke (center), and 1234 sampling days encompassing the March–Oct. "transport season" for more than a decade (right columns). Percentages are for the transport season. Zero indicates that regression cannot resolve a positive contribution to mean concentration, although measurable mass is present in some samples. These results confirm that regional aerosols are dominated by the dusty Asian plume and aged, salt-exposed biomass smoke. Inclusion of days not strongly associated with Asian dust (right) allows detection of local dust. Mean Asian aerosol mass is somewhat less than in Table 2 due to removal of smoky days (left, center) and inclusion of low-mass transport events not detected by the dust recognition algorithm (right). Partial components regressed independently; sums of fractions may not equal totals. Statistics are mean and standard deviation of the population of regression-estimated partial concentrations. Note that for the inclusive transport season case, the 2.5 μm cut approximates the mass median diameter.

of the "pure" aerosol. The statistics presented in Table 2 for the low-smoke/high-confidence Asian dust subset thus can also be seen as characterizing the long-term composition of the dusty Asian aerosol, albeit at a slightly higher concentration than seen on average in the transport season case (Table 3, right). While the dust is the bulk of the coarse fraction, it is important to recognize that it is about one half of the total aerosol, and only about one fourth of the fine material, which is dominated by carbonaceous species. The data indicate that the IMPROVE 2.5 μm cut point falls near the mass median diameter for this aerosol.

6.4. Characteristics of the "Smoky Oceanic Aerosol"

[45] Although the strength of the aged smoke signature is independent of the dusty Asian aerosol, it is strongly linked to days when the dusty aerosol is present. Statistical independence alone does not preclude at least a partial Asian origin. Single-day analyses for the strongest events associated with PC2 in the whole transport season data set are generally associated with the wildfire season in the southern cascade region (August–October). High-PC2 days with very high fine particle concentrations (>10 $\mu g/m^3$ fine carbon) appear to represent combinations of oceanic air masses and regional fires. Back trajectories (HYSPLIT4, available at http://www.arl.noaa.gov/ready/hysplit4.html, 1997) for these peak events indicate North Pacific, Siberian, or Arctic air mass associations. However, the more numerous moderate concentration events (PC scores from 1 to 3 σ above the component mean) show a temporal correlation with the dusty Asian aerosol, implying a link to Asian transport meteorology. In addition, the chemical evidence strongly suggests transoceanic transport. The relationship between the $C/SO_4^=$ ratio and sodium concentrations indicates that reacted sea salt (inferred as $Na + NO_3$) is greatest for events with less than ~10 $\mu g/m^3$ total fine carbon and $C/SO_4^=$ ratios below 10. Comparing this with the distributions in Figure 9, a "first guess" is that the long-range transported smoke is confined to the set of samples with $C/SO_4^= < 6$, while the strong local fire events (high in carbon, low in sodium) populate the high end of the distribution. Unfortunately, the strong chemical similarity of biomass smoke from a variety of sources precludes using the present analyses to further resolve the sources of the smoky oceanic aerosol.

7. Conclusions

[46] 1. The data confirm the initial proposition that these montane sites, remote from major urban areas, are exposed to strong Asian aerosol influences.

[47] 2. The dusty Asian continental aerosol plume is a regular component of the troposphere over western North America with a relatively stable chemical composition (that is, it is statistically separable from other sources). Its typical

concentration is ~6 μg/m3 and mass median diameter is between 2 and 3 μm. The fine fraction (PM2.5) is ~28% soil, 24% carbon, 11% sulfate, and 3% nitrate.

[48] 3. There is also an independent aged, ocean-exposed biomass smoke aerosol that frequently accompanies the dusty Asian aerosol. When present, its typical concentration is ~1.5 $\mu g/m^3$. Evidence linking this aerosol to Asian sources is not definitive in the data reported here; however, a similar aerosol was observed in Asian air masses flowing over the western Pacific in aircraft sampling during the PEM-West B field program [*Moore et al.*, 2003].

[49] 4. Typical coastal marine aerosol signatures are weak or absent at Crater Lake and Mount Lassen. Upslope airflow containing smoke, reacted sea salt, and soil are accounted for and shown to be minor components of the ambient aerosol at these sites. This indicates that the meteorological processes that isolate the montane sites from the marine boundary layer [*VanCuren and Cahill*, 2002] are strong throughout the transport season.

[50] 5. The Asian continental aerosol signature's strong statistical presence in a decade of samples from multiple sites suggests that the Asian plume's previously reported episodic nature is a function of insensitive measurement methods and observational focus on large transport events, rather than a real feature of the atmosphere. Support for this interpretation can be found in a modeling study [*Yienger et al.*, 2000] that found that CO from Asian sources arrives nearly daily in North America. Since Asian materials should be well distributed over the northern Pacific, and allowing for holdover from strong transport days, it is reasonable to expect Asian aerosols to be present, at least in small concentrations, throughout the transport season.

[51] 6. The dominance of Asian aerosols at Crater Lake and Mount Lassen is not geographically confined. The previously reported broad spatial distribution of Asian dust [*VanCuren and Cahill*, 2002] and the wide distribution of the ''baseline'' concentrations for lead and sulfate shown here suggest that the Asian continental aerosol plume is a significant component of long-term aerosol budgets at remote elevated sites across North America.

[52] 7. There is a strong statistical association between dust and combustion products in the Asian continental aerosol plume. (This is not to say that the relative amounts of dust and combustion products are fixed, only that they are closely linked.) The strong association indicates that these materials probably have common source areas and similar temporal patterns of emission. This belies assignment of the bulk of the soil elements to dust storms or other infrequent events. It may be possible that dust from the deserts gets mixed with anthropogenic aerosols as it passes over eastern China, but the reported prevalence of horizontal isentropic transport for dust events [*Wang et al.*, 2000; *Merrill et al.*, 1989] argues against strong vertical motion to mix coastal aerosol with desert dust, and downward movement of dust to the coast would expose it to rapid depletion due to marine deposition. The in-transit mixing explanation also is challenged by the fact that the dusty Asian aerosol, from a mass perspective, is in fact a dust-contaminated combustion aerosol (assuming a factor of 1.4 to convert OC to organic mass, the sum of carbonaceous aerosol, sulfate, and nitrate in the fine fraction of the dusty Asian aerosol is 3.5 times the fine soil). The more probable explanation is that much of the dust, at least for the more numerous low-concentration events, originates from ongoing human activities, including agriculture, transportation, and industry. Support for this interpretation comes from recent dust studies conducted in China that have shown that the majority of dust observed on the Loess Plateau (downwind of the great Chinese deserts) is locally generated [*Zhang et al.*, 1993, 2002; G. Qingxian, personal communication, 2002]. Moreover, the local aerosol measured in heavily polluted Chinese cities [*He et al.*, 2001; *Zhang et al.*, 2002] is chemically similar to the dusty Asian aerosol reported here.

[53] In summary, the data presented here indicate that the Asian continental aerosol plume is a prominent feature of the midlatitude troposphere over North America, and that it has a largely anthropogenic origin. Further research is needed to better understand the particular Asian sources that produce this material and how human activity or climate change may modulate the flux of these aerosols on regional to hemispheric scales.

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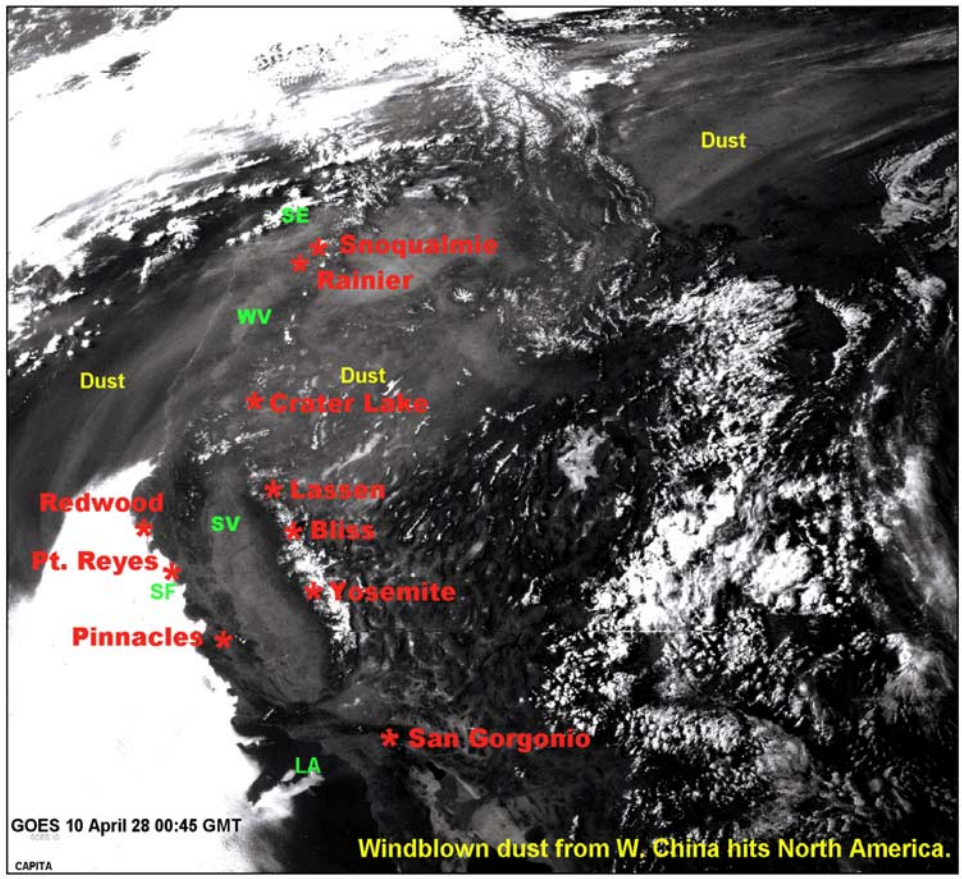

Figure 1. Geostationary Operational Environment Satellite 10 (GOES 10) visible band image of an Asian dust cloud passing over western North America. Stars are coastal and western cordillera IMPROVE sites discussed in text. Marine inversion is marked by low stratus clouds shrouding central and northern California coast; snow cover highlights Sierra Nevada crest and volcanic peaks of the Cascade Range. SE, Seattle; SF, San Francisco; LA, Los Angeles; SV, Sacramento Valley; WV, Willamette Valley (image modified from *Husar et al.* [2001]).

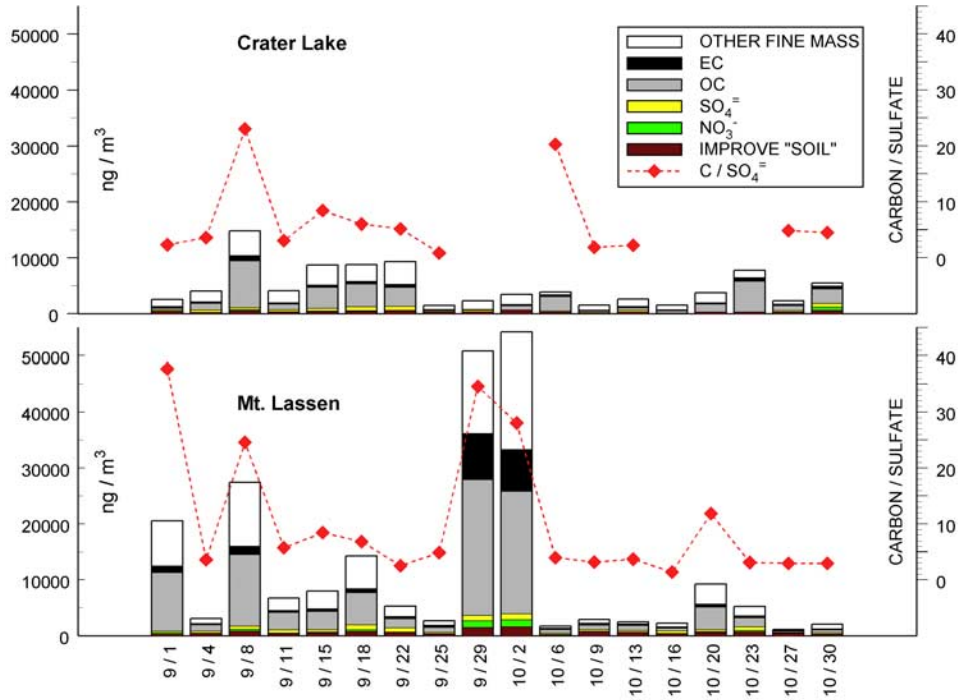

Figure 8. Time series fine aerosol major chemical species at Crater Lake and Mount Lassen, 1 September–30 October 1999. Eastern Pacific air masses with typical fine aerosol burdens of 2–6 μg/m^3 are interrupted by occasional wildfire smoke impacts. The carbon mass change dominates the smoke events, so smoke events exhibit characteristically high carbon/sulfate ratios. Low-smoke days show a lower bound carbon/sulfate ratio in the range of 3 to 6; smoky days regularly exceed 20 (Crater Lake carbon data missing for 29 September; sulfate missing for 2 October).

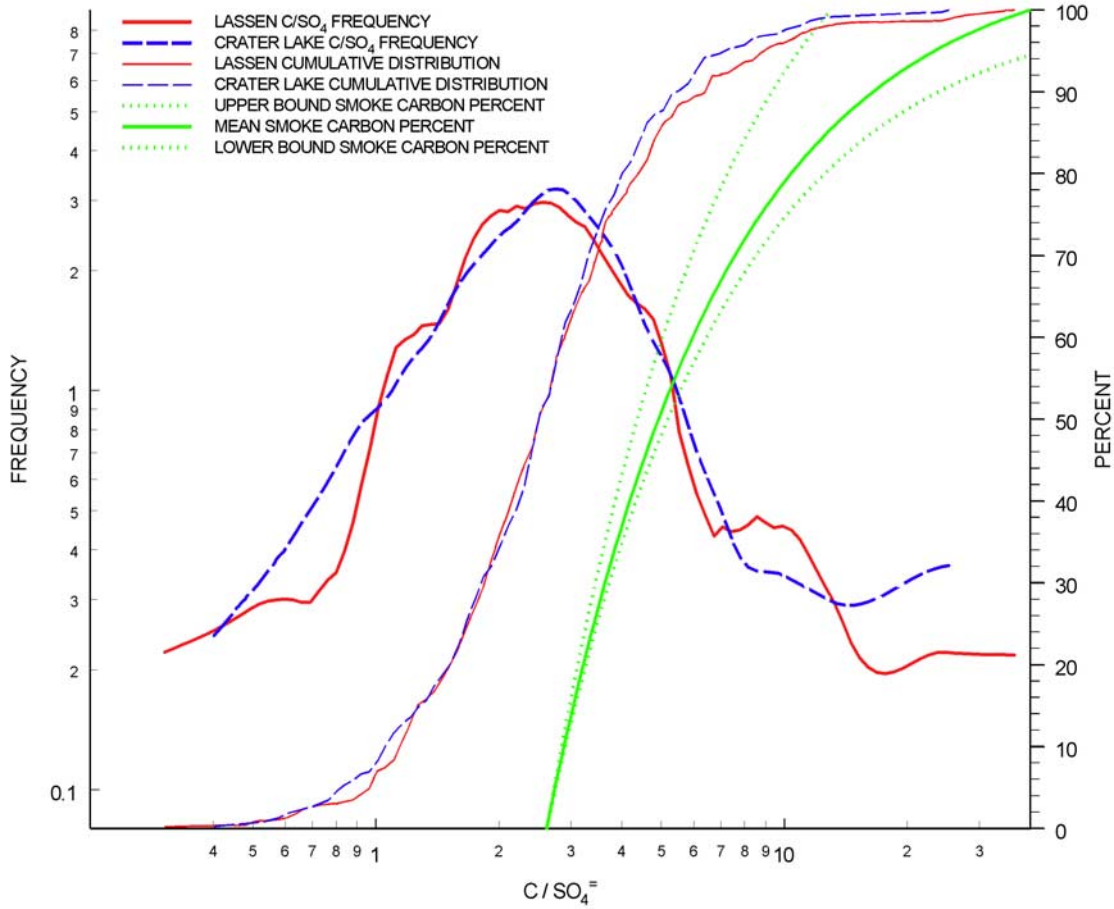

Figure 9. Frequency distributions of $C/SO_4^{=}$ ratios for days with high-confidence Asian dust scores at Crater Lake (258 days) and Mount Lassen (349 days) from 1989–1999. The low to moderate smoke mode ($C/SO_4^{=} < 6$) contains ~90% of the samples. Two smoke-influenced modes are also present, one in the range 6 to 15 (92nd to 97th percentiles), and a second above 20 (top 1% of events). The solid and dashed lines rising from the center show smoke carbon fractions calculated using a simple mixing model assuming the dusty Asian aerosol has a $C/SO_4^{=}$ ratio of 2.7 (the mode of the distribution) and the overall minimum, maximum, and mean smoke ratios from the line ''smoke geometric mean'' in Table 1. Smoke is generally less than half of the carbon and less than about a quarter of total fine mass in the sample population with ratios below 5. Analyses reported for ''low-smoke'' samples used pooled statistics for all samples below 4 as a compromise between capturing the range of Asian aerosol composition and excluding influence from smoke.

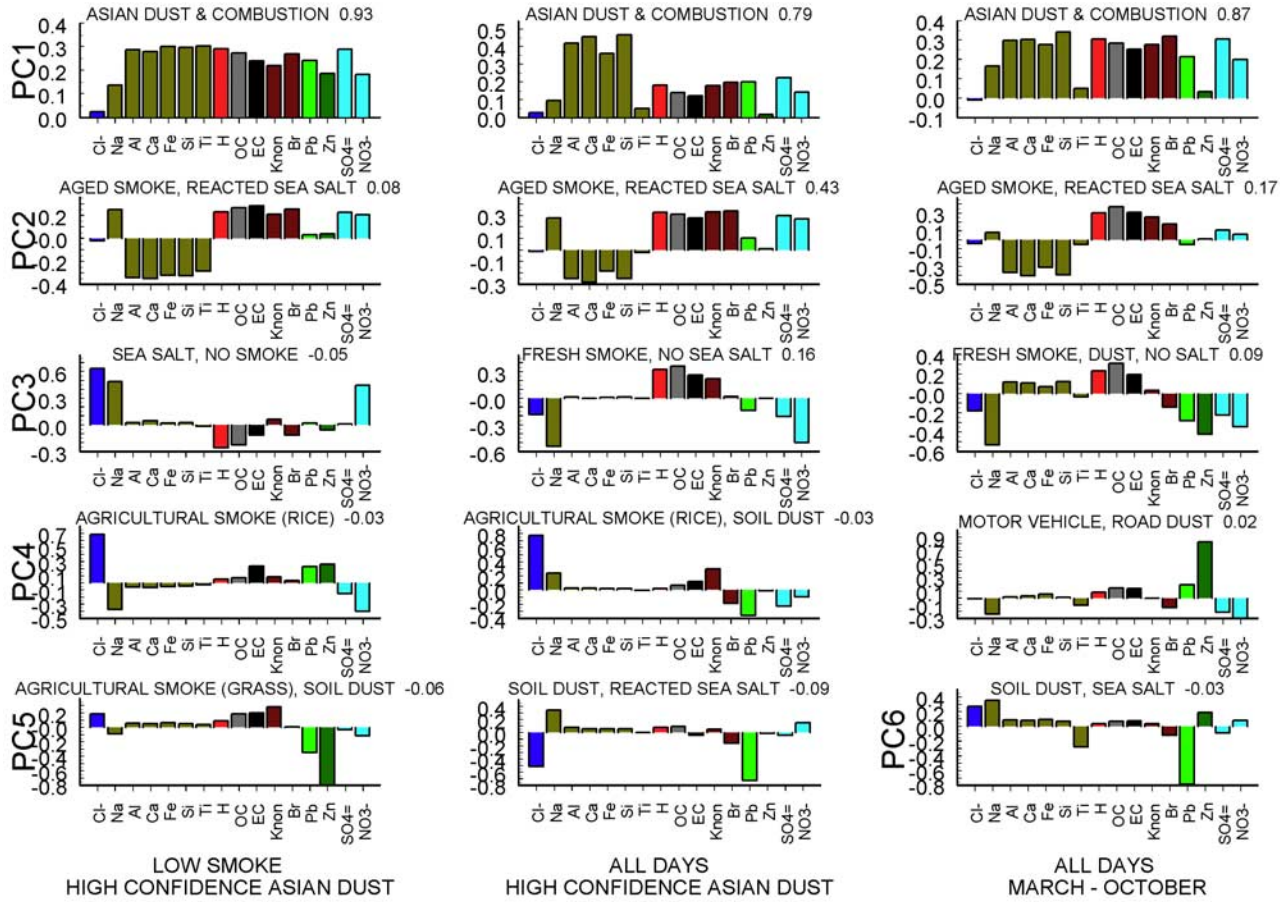

Figure 10. Leading Principal Components (PCA) of pooled IMPROVE data for Crater Lake and Mount Lassen 1989–1999. Left panels show 392 sample days with low smoke ($C/SO_4^{=} < 4$) and high-confidence Asian dust by method of *VanCuren and Cahill* [2002]. Middle panels show 501 sample days with high-confidence Asian dust. Right panels show 1234 sample days in the transport season (March–October). Components computed from measurements transformed to z scores (units of σ) to balance sensitivity among dominant and trace species. The PCs express the covariance structure of the correlation matrix to show which species vary together. Bars represent strength of covariance, not relative mass. Label on each plot gives subjective source category identification and the correlation coefficient of the component with fine aerosol mass. The dusty Asian plume (PC1) and aged smoke with sea salt (PC2) are dominant across all data sets. Agricultural burning and sea salt signify upslope transport. Moving from the selected dust data to dust with smoke to the whole transport season data set there is no new major category associated with the non-dust-identified days (more than half of all days), but weak local sources (fresh smoke, local dust, and vehicle exhaust) rise in importance. The transport season case includes many low-aerosol concentration days associated with the dusty Asian aerosol; thus mass correlation increases even with the strict dust constraint removed. (Transport season PC5, not shown, is an artifact due to nondetection of Zn on low aerosol mass days.) A regression-based mass model for these data is presented in Table 3.